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On the annihilation Doppler effect in various materials

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With 6 figures in the text

It is established through many experiments that positrons penetrating into some solid or liquid material in general slow down to energies of the order of electron volts before annihilation; but there is some doubt whether they end their life as free particles or bound in positronium atoms. It is obvious that the positron when wandering in matter will have the highest probability of finding itself as far away as possible from the positive nuclei, and consequently it may be expected to annihilate most frequently with valence electrons. That is indeed the fundamental result of the investigations that have been made of the momentum distribution of annihilating electron pairs. Up till now one of the following three methods have been employed in such experiments.

Du Mond *et al.* [1] were the first to observe this effect. In recording, with a curved crystal spectrometer, the annihilation line of Cu^{64}, they discovered that the line was somewhat broader than the instrument window curve, calculated from the geometry of source holder and collimator and empirical data on the diffraction properties of the crystal. A rough analysis of the broadening gave a mean momentum of 1.1 (mc/137) for the annihilating pairs, and in a later experiment 1.35 (mc/137).

The motion of the electron pairs manifests itself not only as a Doppler effect but also as a broadening of the 180° correlation between the annihilation quanta. If the momentum component of the electron-pairs perpendicular to the direction of observation is p_z, the deviation φ from 180° becomes

$$\varphi = \frac{p_z}{\mathrm{mc}}$$

for small φ. From the counting rate $N(\varphi)$ the statistical distribution $W(p_z)$ can be deduced, the exact relation depending on the geometrical details of the experiment. In the last few years extensive measurements have been made by this method [2, 3, 4, 5] chiefly on the annihilation in metals. The results show that the momentum distribution is roughly what one expects to find in the Fermi gas of the conduction electrons, which indicates that the positrons reach very low velocities, possibly thermal, before annihilating and that the positron seldom penetrates into the core to annihilate with a bound electron. Investigations of insulators, on the other hand, have yielded strange momentum curves, the shapes of which have not yet been fully understood.

Hedgran and Lind [6] have used the third possible method, viz. analysis of the annihilation photo-line by means of a magnetic β-ray spectrometer. Their work contains only one investigation of this kind, namely that of the annihilation in metallic copper. As the expectation value of the momentum these authors report $\overline{p} = 0.52$ (mc/137), which has to be compared to 1.1 (mc/137) from Du Mond's data and 1.2 (mc/137) from an angular correlation measurement on Au, that has been shown to be almost equivalent to Cu. From the latest report [3] on Cu one calculates from the angular distribution curve the figure $\overline{p} = 1.15$ (mc/137). In spite of the convincing support for the larger value of $\overline{p}$ from several independent measurements, one may object that in principle the photo-electron method ought to be the suitable one for a determination of $\overline{p}$. While the resolution of the crystal spectrometer is barely sufficient and its window has apparently to be computed in a rather delicate way, and while in the coincidence technique one necessarily has to calculate the resolution the method of β-spectroscopy is free from all these objections. Mono-energetic reference lines can easily be found, the size of the sample does not enter into the analysis, and no disturbing effect is caused by different stopping power of positrons in different materials. In order to explain the astonishing discrepancy in annihilation momentum, a redetermination has here been made for Cu^{64}, and in addition CuO and Al have been investigated.

The experiment

The instrument used was a double-focusing spectrometer, $\varrho = 50$ cm, with iron poles. All runs were taken at a solid angle of $2 \times 10^{-3} \times 4\pi$ and at a source and slit size of 2×15 mm^2. The essentials of the source-holder are seen in reference [6], page 33. In the experiments on Cu and CuO, the samples were irradiated for 2 days in the reactor of AB Atomenergi and enclosed in a copper capsule in order to absorb direct β-radiation. Doubtless the best method for obtaining annihilation in materials other than copper compounds would be to mix them in form of a powder with a carrierfree positron emitter. Because of the difficulty of preparing such samples it was preferred in this case to mix, in known proportions, fine Cu and Al powder and to pack the mixture in an Al capsule, which was neutron-irradiated. This technique is particularly useful in the case of Al, which is not observably active after irradiation. In the analysis it was assumed, that the metals were homogeneously blended, that the positrons had a sufficiently small ($< 10\%$) probability of annihilating in the copper powder from which they originated, and that wall effects could be neglected. Homogeneity and grain size were examined under a microscope and it was found that the grains had the form of rather thin shavings with a diameter of some 10 μ. A theoretical estimate of the wall effect in combination with the above measurement, secures the fulfillment of the necessary conditions at the geometry used (the inner diameter of the capsule was 6 mm). Besides, a possible deviation from proportionality to the advantage of Cu would yield a line profile, representing some mean between the ideal Cu and Al curves, and would accordingly only slightly decrease the difference. It was possible to pack about 625 mg powder into the capsule, this amount consisting of 21% Cu, which produced an initial activity of about 60 mc. As a γ-converter was used a 0.1 mm Al foil covered with 1 mg/cm^2 uranium oxide layer, prepared chemically. As the presence of certain inhomogenities in the foil could not be definitely excluded, care was taken always to use the same part of the area.

For the interpretation of the Doppler broadening it is of course important to have an accurate konwledge of the instrument window, that is the line shape corresponding

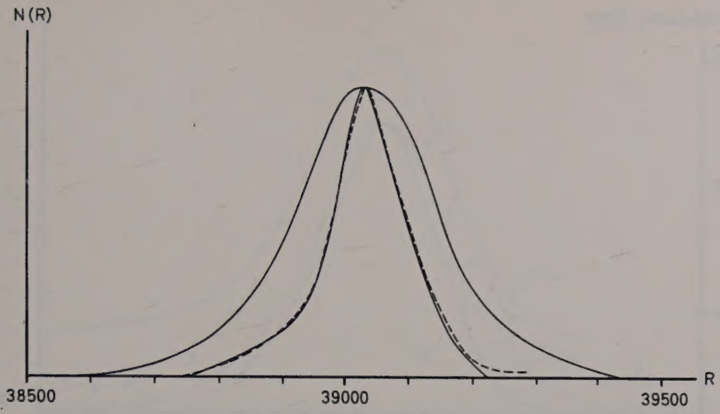

Fig. 1. The annihilation line of metallic Cu, compared with the window curves.

to a sharp γ-line. Hedgran and Lind in their experiment made use of the $U_{L_{III}}$ line of Au^{198} the energy of which coincides almost exactly with the U_K line of the annihilation quanta. In view of the weak intensity of this L line (about 5% of the corresponding K line) another possibility was chosen. The U_K lines of the strongest transitions in As^{76} and Ir^{192} are situated 49 keV above and 43 keV below mc^2 respectively, and can be used in combination to establish a monokinetic line profile. Powdered samples of both metals were packed in Al capsules and irradiated in a pile, after which the As line was recorded twice and the Ir line three times. When corresponding runs had been averaged and the background subtracted, the As and Ir curves were normalized to each other vertically and to the $B\varrho$ of the annihilation line horizontally. The results shown in Fig. 1 coincide excellently, and the average can accordingly be accepted with confidence as a window curve.

Three runs, each comprizing 25 points, were taken for the annihilation in Cu. The curve given in Fig. 2 is representative for these measurements. Fig. 1 shows the mean of the runs, with the normalized window curve in the same diagram for easier comparison.

The Al curve, the result of four similar runs, is seen in Fig. 3, and in Fig. 4 it is compared with the Cu profile. The width of the former is definitely less, in agreement with the angular correlation. In the same figure the CuO curve is drawn an average of two 25 points runs, slightly displaced upwards for the sake of clearness. It is noted that Cu and its oxide show no significant differences in line shape, which is surprising in view of the fact that the compound lacks conduction electrons.

Theory

In every volume element of the sample the annihilating pairs may be assumed to show an isotropic momentum contribution because of the strong straggling of the positrons, when they are slowed down from some 100 keV to the eV-region. The Doppler effect of the γ-quanta that hit a given surface element of the uranium foil comes from the electron velocity component in the direction of propagation of the quantum. Because of the isotropy the integrated result is that

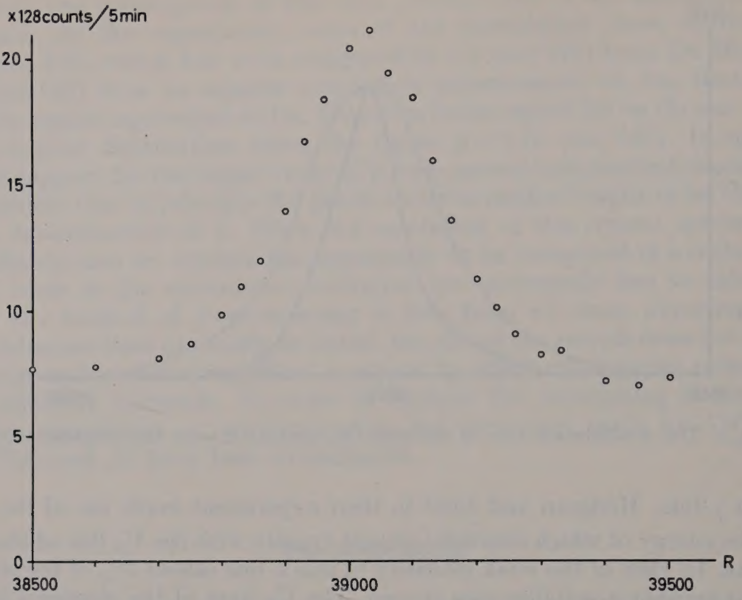

Fig. 2. One of the runs of the annihilation line from Cu.

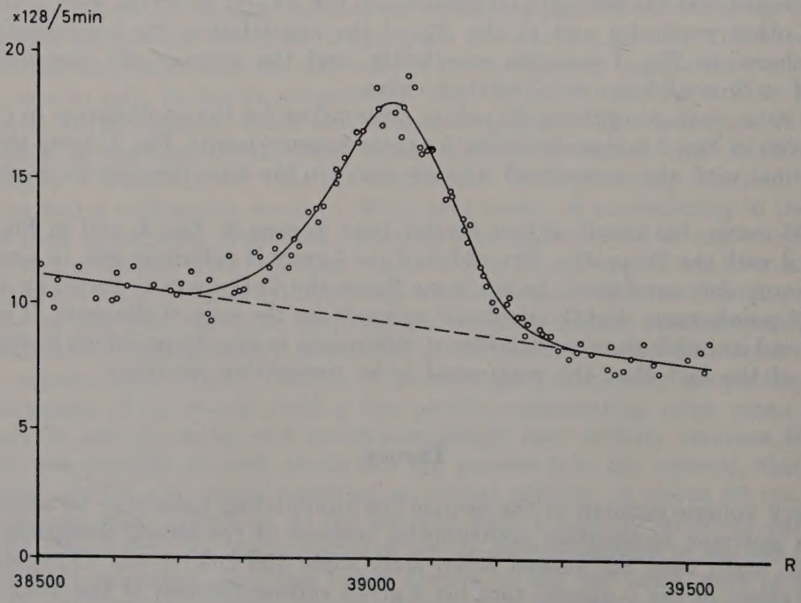

Fig. 3. Experimental points and composite line profile from the runs of the Al–Cu mixture.

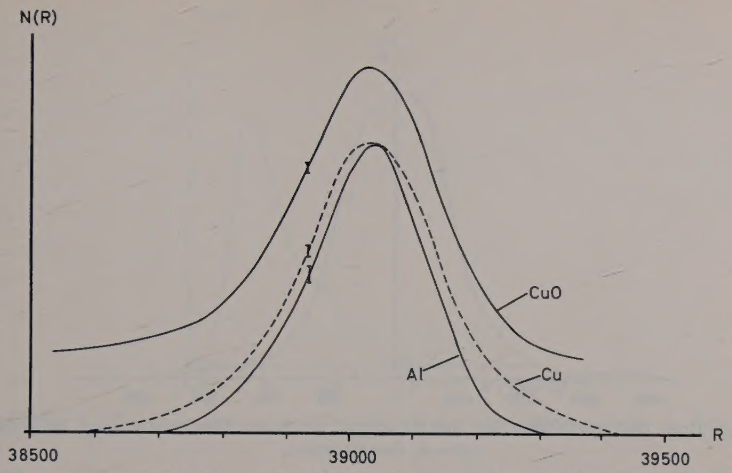

Fig. 4. The line profiles of Al, Cu, and CuO, the last one displaced upward. These curves have been corrected for background and decay but not for the finite resolution.

$$\nu = \nu_0 \frac{1}{1 - \frac{v_z}{c}} = \nu_0 \frac{1}{1 - \frac{p_z}{2\,\mathrm{mc}}} \simeq \nu_0 + \nu_0 \frac{p_z}{2\,\mathrm{mc}}$$

$$\Delta E = h\nu - h\nu_0 = \frac{p_z}{2\,\mathrm{mc}} E_0$$

yielding

$$p_z = 2\,\mathrm{mc} \frac{\Delta E}{E_0},$$

where ν is the frequency in the coordinate system of the converter, ν_0 and E_0 the frequency and energy referred to the center of mass of the electrons, of which p_z is a component of momentum. The above formulae are unrelativistic, which is justified at these moderate velocities. The probability $W(p_z)$ for p_z is consequently proportional to $N(E)$, i.e. to the number of quanta of the corresponding energy. This means that the annihilation line would represent $W(p_z)$ directly if the window curve were sufficiently sharp. As it is actually of comparable width, one has to correct for the finite resolution. If the experimental curve is $f(R)$, R being the potentiometer reading, the window curve $F(R)$ and the corrected line profile $\omega(R)$, one obtains

$$f(R) = \int_{-\infty}^{+\infty} \omega(x)\, F(R-x)\, dx,$$

f being a so-called folding of ω and F. Our problem is to solve ω from the above integral equation. No analytical method of solution exists and one has to use numerical techniques. For this purpose we constructed an iteration process that converged rapidly. As the 0th approximation we chose $f(R)$, as the 1st approximation:

$$f(R) - \left[\frac{\int_{-\infty}^{+\infty} f(x)\, F(R-x)\, dx}{\int_{-\infty}^{+\infty} F(x)\, dx} - f(R) \right]$$

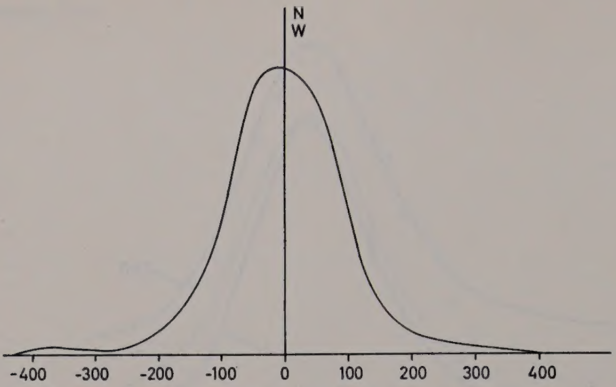

Fig. 5. The Cu line, corrected also for instrumental window. The curve may be interpreted as the p_z distribution.

and so on. Already the 1st approximation was mainly within the experimental errors, but the calculation was carried on in one more iteration, yielding the final curve in Fig. 5. The resulting symmetry, which was not introduced as an explicit condition in the numerical process, supports the experimental data.

From $\omega(R)$ we calculate $$\bar{R} = \frac{\int x\,\omega(x)\,dx}{\int \omega(x)\,dx}$$

which is a direct measure of $\overline{|p_z|}$. It is then easily shown that for an isotropic distribution

$$\bar{p} = 2\overline{|p_z|}.$$

For an isotropic distribution of monokinetic particles we have

$$W(p_z)\,dp_z = \begin{cases} \dfrac{1}{2p}\,dp_z & \text{for } -p < p_z < p, \\ 0 & \text{for } |p_z| > p. \end{cases}$$

From this one immediately deduces the generalization to a distribution $W_p(p)$ of p

$$W(p_z) = \int_{p_z}^{\infty} \frac{1}{2p} W_p(p)\,dp$$

and consequently

$$W_p(p) = \text{const} \times p\,\frac{dW}{dp}.$$

Results

Of the three materials that have been investigated here, copper has been more extensively analyzed than the others. The corrected Doppler curve $\omega(R) = \text{const} \times W(p_z)$ has been calculated and from this function $\bar{p}$ has been determined. Our value

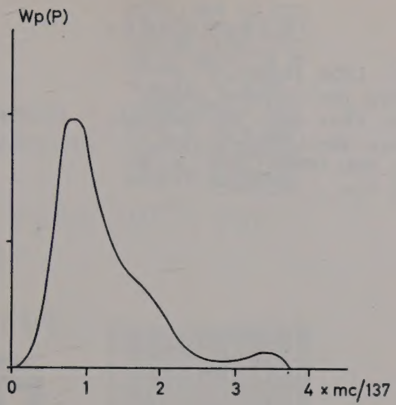

Fig. 6. The momentum distribution of the annihilating pairs in Cu.

$\overline{p} = 1.3$ (mc/137) is more than twice that obtained previously by Hedgran and Lind. Their paper, however, contains no thorough discussion of the physical background of the annihilation experiment, and it might be feared that they have not made the necessary distinction between $\overline{p}$ and $\overline{|p_z|}$, in which case their result should be interpreted $\overline{p} = 1.04$ (mc/134). This value is still somewhat lower than ours, but it is doubtful whether the difference should be given any significance[1]; in any case the errors overlap. The essential result is that the previously existing discrepancy has been removed and the three different methods of investigating annihilation momentum have been shown to give equivalent results.

In Fig. 6, $W_p(p)$ gives a more detailed picture of the momentum distribution. One notices especially the pronounced high energy tail, which has previously been found in Zn by angular correlation [5] but which is absent in lighter metals for instance in Mg. The effect is no doubt due to positrons penetrating into the region of bound electrons. Most of the annihilations are, however, thought of as occurring with the conduction electrons, the velocity distribution of which can be found independently. One may expect that a careful measurement of $W_p(p)$ might throw some light on the possible existence of positronium in metals, since a two-step process of annihilation would yield a different momentum distribution than the direct one.

I wish to thank Professor Kai Siegbahn for suggesting an investigation on annihilation Doppler broadening and for his most stimulating interest in this work. I also wish to acknowledge the kind assistance of Miss B. Lindqvist and Mr. G. Lindblom in operating the spectrometer.

Uppsala, May 1956.

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[1] The most plausible explanation is that our measurements do more justice to the high energy tail.

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